PB82-132481

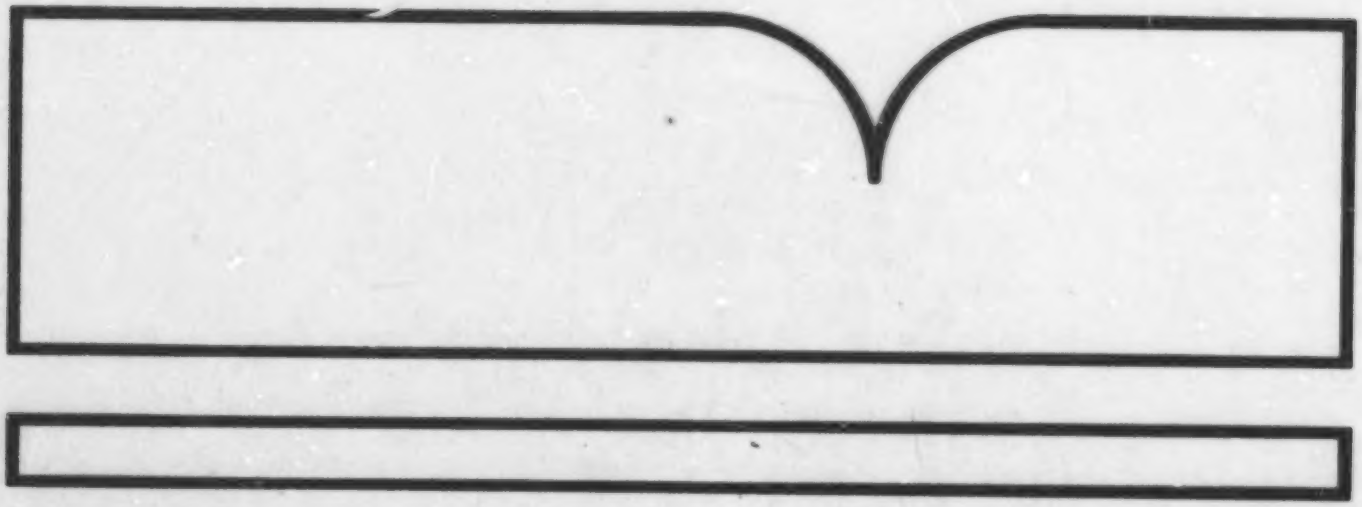
A Determination of the Sources of Airborne Particles Collected during the Regional Air Pollution Study

Illinois Univ. at Urbana-Champaign

Prepared for

Environmental Sciences Research Lab.
Research Triangle Park, NC

19 Jun 80



U.S. Department of Commerce
National Technical Information Service
NTIS

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA-600/J-81-025	2. Journal Article
3. RE PB82-132481	
4. TITLE AND SUBTITLE A DETERMINATION OF THE SOURCES OF AIRBORNE PARTICLES IN AEROSOL SAMPLES COLLECTED DURING THE REGIONAL AIR POLLUTION STUDY	5. REPORT DATE November 1981
	6. PERFORMING ORGANIZATION CODE 2337067
7. AUTHOR(S) Daniel J. Alpert and Philip K. Hopke	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Institute for Environmental Studies and Nuclear Engineering Program University of Illinois Urbana, Illinois 61801	10. PROGRAM ELEMENT NO. CDTA1D/09-0357 (FY-81) CDTA1D/09-0436 (FY-81)
	11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS Environmental Sciences Research Laboratory, RTP,NC Office of Research and Development U. S. Environmental Protection Agency Research Triangle Park, North Carolina 27711	13. TYPE OF REPORT AND PERIOD COVERED
	14. SPONSORING AGENCY CODE EPA/600/09

15. SUPPLEMENTARY NOTES

16. ABSTRACT

Target transformation factor analysis (TTFA) has been applied to an analysis of a subset of the aerosol-composition data acquired during the Regional Air Pollution Study (RAPS) for St. Louis, Missouri. The RAPS program collected a large number of samples with ten, continuously operated, dichotomous samplers from March 1975 to March 1977. The purpose of the present study was to evaluate the capability of TTFA to resolve sources of airborne particulate matter in a set of ambient-aerosol samples. In order to give each element a more equal weight in the identification of sources, a weighting scheme has be n added to the target transformation rotation procedure. The weighted rotation produces a more sensitive source identification and has enhanced the resolution of sources with similar element profiles. To determine the most appropriate way to apply TTFA, two separate sets of the data were analyzed: 1) All the samples collected during July and August 1976 at a single station, and 2) All the samples collected at the ten RAPS stations during a single week. Each set of samples were further subdivided into fine- and coarse-faction subsets and analyzed separately. Because of the large number of missing values below detection limits, it was necessary to exclude a sizable fraction of the data from the analyses. Superior results were obtained from the examination of the variation in aerosol composition with time at a single location rather than the spatial variation over multiple sites during a shorter time period.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS *(This Report)* UNCLASSIFIED	21. NO. OF PAGES
	20. SECURITY CLASS *(This page)* UNCLASSIFIED	22. PRICE

EPA Form 2220-1 (9-73)

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA 22161

EPA-600/J-81-025
Journal Article

Atmospheric Environment Vol. 15, No. 5, pp. 675–687, 1981
Printed in Great Britain.

0004–6981/81/050675–13 $02.00/0
© 1981 Pergamon Press Ltd.

A DETERMINATION OF THE SOURCES OF AIRBORNE PARTICLES COLLECTED DURING THE REGIONAL AIR POLLUTION STUDY

DANIEL J. ALPERT* and PHILIP K. HOPKE†
Institute For Environmental Studies and Nuclear Engineering Program, University of Illinois, Urbana, Illinois 61801, U.S.A.

(*First received* 5 *May* 1980 *and in final form* 19 *June* 1980)

Abstract—Target transformation factor analysis (TTFA) has been applied to an analysis of a subset of the aerosol-composition data acquired during the Regional Air Pollution Study (RAPS) for St. Louis, Missouri. The RAPS program collected a large number of samples with 10 continuously operated dichotomous samplers from March 1975 to March 1977. The purpose of the present study was to evaluate the capability of TTFA to resolve sources of airborne particulate matter in a set of ambient-aerosol samples. In order to give each element a more equal weight in the identification of sources, a weighting scheme has been added to the target transformation rotation procedure. The weighted rotation produces a more sensitive source identification and has enhanced the resolution of sources with similar element profiles. To determine the most appropriate way to apply TTFA, two separate sets of the data were analyzed: (1) all the samples collected during July and August 1976 at a single station; and (2) all the samples collected at the ten RAPS stations during a single week. Each set of samples was further subdivided into fine- and coarse-fraction subsets which were analyzed separately. Because of the large number of missing values and values below detection limits, it was necessary to exclude a sizable fraction of the data from the analyses. Superior results were obtained from the examination of the variation in aerosol composition with time at a single location rather than the spatial variation over multiple sites during a shorter time period.

INTRODUCTION

In 1971, the first National Ambient Air Quality Standards were established, setting limits on permissible levels of total suspended particulate matter. Because these regulations concentrated only on the absolute amounts of suspended particulate matter, control efforts were directed primarily toward the removal of the larger, more massive particles that comprise most of the total mass of particle emissions. There is growing evidence, however, that the smaller particles, although representing a smaller fraction of the total mass, present a greater threat to public health and welfare (Commission on Natural Resources, 1979). In addition, the smaller particles are believed to contribute to a larger extent to the reduction in visibility. New regulations, now under review by the EPA, may include standards not only on the mass of total suspended particles but on the levels of inhalable (diameters less than 15 μm) and fine (diameters less than 2 μm) particles as well. The effectiveness of regulations designed to limit the impact of fine and inhalable particles on the public would be greatly enhanced if the sources and relative emission rates of these particles were known. Because of the complexity of urban ecosystems, determining the sources of airborne particulate matter is a very difficult problem. The problem is exacerbated by the presence of fugative sources and the lack of understanding of the modes of formation and transport of airborne particulate matter. The advent of receptor methodology and recent developments in the areas of trace-element analysis and multivariate statistical techniques now permit a much more detailed analysis of ambient aerosol samples. By providing detailed information on the sources of fine and inhalable particles, such techniques could become a major part of any strategy for controlling airborne particulate matter.

The majority of recent source-resolution work has used the chemical element balance method (CEB) developed by Miller, Friedlander, and Hidy (1972). In this method, it is assumed that the number and composition of the sources are known. The observed elemental concentrations in a set of ambient aerosol samples are fit by means of a multiple regression analysis. CEB has produced very good fits to the data in recent studies of Washington, D.C. (G. Kowalczyk, 1979) and St. Louis, MO (Dzubay, 1980). Watson (1979) greatly expanded the scope of the chemical element balance method in his study of the Portland, OB airshed. In that study, the element profiles of approximately 30 different sources were measured. By including a much larger number of source components, very good agreement was achieved for the 23 elements and chemical species that were included in the analysis. Chemical element balance methods, however, have a major drawback in that they require an *a priori* knowledge of both the number and composition of the suspected source emissions. The only quantities calculated are the contributions of each of the presumed

* Current Address: Division 4413, Sandia National Laboratories, Albuquerque, NM 87185, U.S.A.
† Author to whom correspondences should be addressed.

sources to each of the measured samples. Though extensive efforts have produced very good determinations of the element profiles of a number of sources, these determinations are time consuming, expensive, and, in many cases, inapplicable to the resolution of sources in different locations.

Another form of statistical analysis, factor analysis, has also been used to identify sources of airborne particulate matter. The major advantage of this method is that it requires no prior assumptions about the nature of the system under study. Factor analysis has been used to identify sources of airborne particulate matter in Boston, MA, (Hopke *et al.*, 1976), Tucson, AZ (Gaarenstroom *et al.*, 1977), and St. Louis, MO (Gatz, 1978). Because of the nature of the factor model used in these applications, the actual mass contribution of each of the identified sources to each sample could not be determined. Several recent studies (Hopke *et al.*, 1980; Alpert and Hopke, 1980b) have employed a new form of factor analysis called target transformation factor analysis (TTFA). Originally developed by Weiner and co-workers (1970), TTFA permits an analysis, similar in nature to the chemical element balance method, but without the presumptions as to the number or nature of the sources in the system. Unlike the earlier applications of factor analysis, TTFA allows the direct calculation of the contribution of each source to each sample. This paper presents an analysis of particulate samples collected as part of the Regional Air Pollution Study (RAPS) of St. Louis, MO using target transformation factor analysis. By providing reliable quantitative source resolutions, TTFA could be an important tool in the development of cost-effective strategies for the control of airborne particulate matter.

TARGET TRANSFORMATION FACTOR ANALYSIS

The fundamental assumption of factor analysis is that the amount of each element in each aerosol sample can be represented by a sum of the contributions from each of the sources that comprise the system. This model is of the form

$$x_{ij} = \sum_{k=1}^{p} a_{ik} f_{kj}$$

where x_{ij} is the amount of element i in sample j and p is the number of independent sources. The contribution from each source is represented as a product of two cofactors, a_{ik} and f_{kj}. The former represents the concentration of the ith element in the particles emitted by the kth source and the latter is the amount of particulate matter per unit volume contributed by source k to sample j. The entire set of aerosol samples can be resolved into source contributions given by the matrix equation

$$X = AF.$$

The objectives of a factor analysis are (1) to determine p, the number of independent sources that contribute to the system; (2) to determine the components of matrix A, the elemental-source profiles; and (3) to calculate F, the mass contribution of each source to each sample.

Details of target transformation factor analysis have been fully described in previous publications (Hopke *et al.*, 1980; Alpert and Hopke, 1980a and 1980b). Briefly, the number of sources is determined by performing an eigenvalue analysis on the matrix of correlations between the samples. A target transformation analysis (Weiner *et al.*, 1970; Malinowski and Howery, 1980) is used to identify the nature of the sources. Target transformation determines the degree of overlap between an input source profile and one of the calculated factor axes. The input source profiles, called test vectors, are developed from existing knowledge of the emission profiles of various sources. Unique test vectors, having values of all but one element equal to zero are also analysed. The uniqueness test can resolve sources that are identified primarily with only one element. To give each element in the test vector an equal weight in the target transformation, a standard least-squares weighting scheme has been included in the analysis (Clifford, 1973). The weighted rotation has greatly enhanced the ability of the target transformation to resolve sources with similar concentration profiles (Alpert and Hopke, 1980a). When a complete set of plausible sources has been isolated, they are scaled to reflect the actual concentration of each element in the source material. Correct scaling factors for all source profiles are determined by means between a multiple regression between the calculated source contributions and the total measured sample weights (Hopke *et al.*, 1980). The scaled test vectors are then used to calculate the contribution of each source to each sample.

DESCRIPTION OF DATA

In the RAPS program, automated dichotomous samplers (Loo *et al.*, 1976) were operated over a two year period at ten sites in the St. Louis metropolitan area. A map of the St. Louis metropolitan area, showing the location of the ten dichotomous samplers, may be found in Dzubay (1980). Ambient aerosol samples were collected in fine, < 2.4 μm, and coarse, 2.4 to 20 μm, fractions (Nelson, 1979). The samples were analyzed at the Lawrence Berkeley Laboratory for total mass by beta-gauge measurements and for 27 elements by X-ray fluorescence (Goulding *et al.*, 1978). Only the data for the months of July and August 1976 were examined in the present study.

To determine the most appropriate way to apply target transformation factor analysis to a large set of aerosol samples, two subsets of these data were analyzed: (1) all the samples collected during July and August at one station; and (2) the samples collected during a single week at all ten RAPS stations. The data from

station 112 were selected for the first subset and the week beginning July 31, 1976 for the second. The relative completeness of the data in these two subsets was the primary criterion for their selection.

Station 112 was located near Francis Field, on the campus of Washington University, west of downtown St. Louis. During the 62 days of July and August, filters were changed at 12 h intervals producing a total of 124 samples in each the fine and coarse fractions. Data were missing for 48 of the samples leaving a total of 200; 99 in the fine fraction, and 101 in the coarse. Of the 27 elements determined for each sample, a majority of the determinations of 10 elements had values below detection limits. Since a complete and accurate data set is required to perform a factor analysis, these 10 elements were eliminated from the analysis. Unfortunately, arsenic was among the excluded elements. Arsenic data may be important to the differentiation of coal flyash and crustal material: two sources with very similar element profiles. Because of limitations in available computer space, the remaining 200 samples were divided into two separate parts. Two different divisions in the data were made to determine the division that permitted the extraction of the most information. First, the data were divided into groups corresponding approximately to the months of July and August. In the second analysis, the data were divided into groups corresponding to the fine and coarse fractions of both months. A more detailed source resolution was obtained from the analysis of the data divided into particle-size fractions (Alpert and Hopke, 1980a). Only the results of the analyses of the data divided into particle-size fractions will be reported here.

During the week beginning July 31, 1976, filters were changed at 12 h intervals at five of the stations and at 6 h intervals at four stations. There were no data from one station. A total of 182 samples were collected in each of the fine and coarse fractions. Data were missing for 43 samples and another 50 samples had a majority of values below the detection limits; these 93 samples were excluded from the analysis. Of the 27 elements, a majority of the determinations of 12 had values below detection limits. These 12 elements were excluded. Again, two separate divisions in the data were made and each individually analyzed. The data were first divided into two groups composed of days Saturday, Sunday and Monday (SSM), and of days Tuesday, Wednesday, Thursday and Friday (TWTF). The first group, with 115 samples, corresponds to days 213, 214 and 215, and the second group, with 156 samples, to days 216 through 219. The analysis was then repeated with the data divided into groups corresponding to the fine and coarse fractions with 139 and 132 samples respectively. Again, superior results were obtained from the analysis of the data divided by particle-size fractions and only these results will be reported here.

The 27 elements determined for each sample account for only a small fraction of the total sample mass. Figure 1 depicts the composition of the average sample in both the fine and coarse fractions at RAPS station 112. The measured elements account for only about 22% of the average sample mass in the fine fraction and about 43% in the coarse. The low percentage of measured elements can lead to distortions in the scaling factors produced by the multiple regression analysis. The remaining mass consists primarily of hydrogen, oxygen, nitrogen and carbon. Determining the sources of particulate carbon is of growing concern because of the potential mutagenicity associated with carbonaceous particles (Daisey *et al.*, 1978). Measurements have shown that both soot and organic carbon can account for a sizable fraction of the total mass of airborne particulate matter. Lewis and Macias

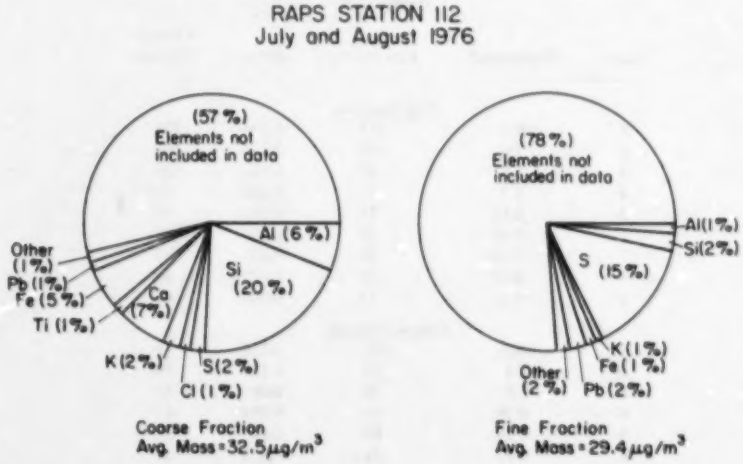


Fig. 1. Composition of the average sample in the fine and coarse fractions during July and August 1976 aT RAPS station 112. Hydrogen, oxygen, nitrogen and carbon constitute most of the mass not included in the data.

(1980) measured a carbon concentration of 16% in the fine fraction in Charleston, West Virginia. A carbon concentration of 37% was reported for Portland, Oregon (J. Kowalczyk *et al.*, 1979). Radiocarbon studies have determined that over 80% of the particulate carbon in the St. Louis aerosol is of fossil-fuel origin (Lodge *et al.*, 1960, as cited by Currie *et al.*, 1978). Though no measurements of carbon are included in the RAPS data, that portion of the sample mass must still be accounted for by the resolved sources. In order to produce the best possible source resolutions, it is vital to have accurate measurements of the mass of total suspended particles (TSP) as well as determinations for as many elements as possible.

RESULTS OF THE ANALYSIS OF SAMPLES FROM STATION 112

The first step in the analysis was to determine the minimum number of factors needed to represent the fine- and coarse-fraction data sets. The results of the tests that aid this determination are given in Table 1. For each number of retained factors, the chi-square value, exner value (Exner, 1966), and average percent error give an indication of the point-by-point error between the original data and the data reproduced with that number of factors. In the fine-fraction data, the large change in the magnitude of successive eigenvalues combined with the low exner value indicated that four factors should be retained. The test results for the coarse fraction were not conclusive, indicating either four or five factors. The nature of the sources in each fraction was determined with a target transformation analysis. In addition to the unique vectors, seventeen different source profiles were tested to determine the degree of overlap with any of the calculated factor axes. The seventeen test vectors included nine different crustal factors, two determinations of coal flyash, three motor vehicle factors, and three industrial sources. In the fine-fraction data, potassium was found to be unique but related to the concentration of Al, Cl, Zn, Sr, and Ba. This factor is attributed to the presence of fireworks particles resulting from the July 4th Bicentennial display in Francis Field. Firework particles were found to affect three samples from July 4th and 5th in the fine fraction and one sample in the coarse fraction. The resolution of a fireworks factor in the fine fraction, though present in only three of the one hundred samples, illustrates the source resolution power of this method. To examine the effects caused by the presence of the fireworks factor, the fine-fraction data were analyzed both with and without all 4 samples from July 4th and 5th. One sample from July 4th was excluded from the coarse fraction. Table 2 compares the average concentration of the elements in the fine fraction both with and without the July 4th samples. Removal of only four of the ninety-nine samples resulted in significant changes in the average concentrations of four elements: Al, K, Sr and Ba. These elements are the primary constituents of the fireworks particles. The results of the dimensionality tests for the fine-fraction data without July 4th and 5th are given in Table 3. There are large changes in the magnitude of the eigenvalues with both three and five retained factors. The exner test indicates three factors while the chi-square test indicates five. Since the tests indicated only four factors when the July 4th samples are included, it appears that in addition to the fireworks factor, there are three major and two minor factors in the fine-fraction data. The two weaker

Table 1. Results of dimensionality tests, RAPS station 112, July and August 1976

Factor	Eigenvalue	Chi square	Exner	Average % error
		Fine fraction		
1	90.0	213	0.324	204
2	5.0	156	0.214	164
3	1.7	65	0.141	129
4	1.3	63	0.064	93
5	0.16	55	0.047	72
6	0.09	26	0.034	68
7	0.03	24	0.027	67
8	0.02	24	0.021	58
9	0.02	15	0.016	49
		Coarse fraction		
1	96.0	202	0.216	73
2	2.4	70	0.125	50
3	0.6	70	0.089	45
4	0.29	59	0.064	41
5	0.19	63	0.040	35
6	0.07	61	0.028	33
7	0.03	38	0.019	28
8	0.02	26	0.013	23
9	0.01	30	0.009	22

Table 2. Comparison of data with and without samples from July 4th and 5th (ng m^{-3}). RAPS station 112, July and August 1976. Fine fraction

Element	With July 4th & 5th Mean	Without July 4th & 5th Mean*
Al	220 ± 30	200 ± 30
Si	440 ± 60	450 ± 60
S	4370 ± 310	4360 ± 320
Cl	90 ± 10	80 ± 9
K	320 ± 130	150 ± 9
Ca	110 ± 10	110 ± 10
Ti	63 ± 13	64 ± 13
Mn	17 ± 3	17 ± 3
Fe	220 ± 20	220 ± 20
Ni	2.3 ± 0.2	2.3 ± 0.2
Cu	16 ± 3	15 ± 3
Zn	78 ± 8	75 ± 8
Se	2.7 ± 0.2	2.7 ± 0.2
Br	140 ± 9	130 ± 8
Sr	5 ± 4	1.1 ± 0.1
Ba	19 ± 5	15 ± 4
Pb	730 ± 50	720 ± 50

* Mean value and standard deviation of the mean.

Table 3. Results of dimensionality tests, RAPS station 112, July and August 1976 (fine fraction 4 and 5 July excluded)

Factor	Eigenvalue	Chi Square	Exner	Average % error
1	87.0	208	0.304	197
2	4.9	152	0.304	197
3	2.0	57	0.070	123
4	0.2	42	0.050	98
5	0.1	26	0.037	73
6	0.04	25	0.029	69
7	0.02	26	0.023	69
8	0.02	17	0.019	67
9	0.01	16	0.015	53

factors were not resolved when the samples from July 4th were included.

The target transformation analysis for the fine fraction without July 4th and 5th indicated the presence of a motor vehicle source, a sulfur source, a soil or flyash source, a titanium source and a zinc source. The presence of the sulfur, titanium and zinc factors was determined by the uniqueness test for those elements. The concentration of sulfur was not related to any other elements and probably represents a secondary sulfate aerosol resulting from primary emissions of sulfur oxide. Titanium was found to be associated with the elements sulfur, calcium, iron and barium. Rheingrover (1977) identified the source of titanium as a paint-pigment factory located to the south of station 112. The zinc factor, associated with the elements chlorine, potassium, iron and lead, is attributed to refuse-incinerator emissions. This factor may also represent particles from zinc and/or lead smelters, though a high chlorine concentration is usually associated with particles from refuse incinerators (Greenberg *et al.*, 1978). With July 4th and 5th included, the results indicated, in addition to the fireworks source, only the presence of motor vehicle, sulfate, and soil or flyash sources. Thus, we conclude that the presence of the fireworks factor inhibits the resolution of the weaker paint-pigment and refuse factors. Based on the degree of alignment with the data, a motor vehicle, paint-pigment, and crustal source were selected from the set of input source profiles. Since the fireworks and sulfate factors had no prior source determinations and the refuse factor differed from that measured by Greenberg *et al.* (1978) in the relative concentration of chlorine, source vectors predicted by the uniqueness test were used for these source profiles. Each of the profiles were refined by replacing the original values for each element with the values predicted by the target transformation.

The absolute concentrations of the test vectors used in the target transformation may not correspond exactly to the source compositions found in the St. Louis area. The predicted source profiles were scaled to the correct absolute concentrations by means of a multiple regression analysis as described by Hopke *et al.* (1980). The calculated source profiles that best reproduced the fine-fraction data are listed in Table 4. The sulfur concentration in the refined sulfate factor is consistent with that of ammonium sulfate. The calculated lead concentration in the motor vehicle factor of 10% with a lead to bromine ratio of about 0.28 are typical of values reported in the literature (Dzubay *et al.*, 1979). The concentration of lead in motor vehicle exhaust will vary from city to city and is dependent on the ratio of leaded gasoline to unleaded and diesel-powered vehicles. The calculated source profile for the refuse factor, with high concentrations of chlorine, zinc and lead, is similar to that measured by Greenberg *et al.* (1978) for the Nicosia Municipal Incinerator near Chicago. However, Greenberg *et al.* (1978) found chlorine, zinc and lead concentrations of 27%, 11% and 7% respectively. In the present study, the chlorine concentration is only 2% and the zinc and lead concentrations are half those found by Greenberg. The lower calculated concentrations may result from the combining of both refuse-incinerator and lead/zinc-smelter emissions into a single factor. In the paint-pigment component, the titanium and iron concentrations are similar to those calculated by Dzubay (1980). There are no previous determinations of a fireworks component but the main ingredients of gunpowder are carbon, sulfur, and saltpeter (KNO_3) so a calculated potassium concentration of 58% seems reasonable. The nature of the calculated soil/flyash factor is more like that of coal flyash than soil, though the absolute concentrations of the major elements are less than those reported by Gladney (1974) and Fisher *et al.* (1978) for coal flyash. Because of the similarity of their elemental profiles, differentiating soil and coal flyash is a problem often encountered in aerosol source resolutions. Coal flyash emissions are expected to contribute more to the fine fraction while crustal material should be found in the

Table 4. Refined source profiles (mg g^{-1}). RAPS station 112, July and August 1976

Element	Motor vehicle	Sulfate	Flyash/ soil	Paint	Refuse
Al	5.0	1.1	53.0	0.0	0.0
Si	0.0	1.9	130.0	0.0	7.0
S	0.02	240.0	19.0	6.0	0.0
Cl	2.4	1.1	0.0	4.6	22.0
K	1.4	1.6	15.0	5.7	48.0
Ca	11.0	0.0	16.0	34.0	1.2
Ti	0.0	0.7	2.5	110.0	0.0
Mn	0.0	0.0	0.7	4.8	8.6
Fe	0.0	1.1	36.0	90.0	36.0
Ni	0.08	0.04	0.042	0.011	0.7
Cu	0.6	0.01	0.0	0.0	8.7
Zn	0.8	0.0	0.0	3.7	65.0
Se	0.1	0.1	0.001	0.2	0.2
Br	30.0	0.03	2.5	0.0	0.05
Sr	0.09	0.01	0.15	0.1	0.005
Ba	0.7	0.05	0.07	28.0	0.5
Pb	107.0	6.5	5.0	0.0	46.0

Table 5. Refined source profiles (mg g^{-1}). RAPS station 112, July and August 1976. Coarse fraction

Element	Soil	Limestone	Sulfate	Paint
Al	71.0	30.0	28.0	5.0
Si	274.0	150.0	0.0	0.0
S	4.9	0.0	90.0	37.0
Cl	1.4	16.0	3.6	6.9
K	19.0	15.0	9.3	0.0
Ca	40.0	188.0	0.08	25.0
Ti	0.0	1.5	0.0	128.0
Mn	0.8	1.6	1.0	1.2
Fe	40.0	34.0	43.0	65.0
Ni	0.01	0.2	0.2	0.06
Cu	0.0	0.6	0.9	0.0
Zn	0.0	4.2	4.3	0.3
Se	0.01	0.02	0.13	0.001
Br	0.5	3.1	3.8	1.3
Sr	0.2	0.3	0.2	0.1
Ba	0.6	0.7	0.4	3.2
Pb	1.5	11.0	13.0	6.7

Fine fraction, including July 4th and 5th

Element	Vehicle	Sulfate	Flyash/soil	Fireworks
Al	3.0	0.9	62.0	60.0
Si	0.0	2.8	140.0	0.0
S	0.0	232.0	14.0	26.0
Cl	5.2	1.6	0.31	19.0
K	0.0	0.06	43.0	580.0
Ca	12.0	0.006	17.0	0.27
Ti	2.8	1.8	2.3	0.0
Mn	1.5	0.1	0.8	3.6
Fe	5.8	3.8	38.0	9.0
Ni	0.2	0.06	0.05	0.3
Cu	1.9	0.2	0.03	4.6
Zn	9.8	1.4	0.0	24.0
Se	0.1	0.1	0.0	0.01
Br	26.0	0.0	2.7	2.0
Sr	0.0	0.0	0.9	12.0
Ba	1.45	0.3	0.8	15.0
Pb	105.0	8.0	3.8	0.0

coarse fraction. Thus, we conclude that this factor is primarily the result of coal-burning power plant emissions. Reliable data for elements, such as arsenic, would be needed to clearly differentiate the contributions of soil and coal flyash.

For the coarse fraction, the target transformation indicated the presence of a sulfate factor, a paint-pigment factor, and two crustal factors; an aluminosilicate type and a limestone or cement source. In an earlier factor analytical study of aerosol sources in the St. Louis area, Gatz (1978) found the element calcium to be associated with other than crustal sources. The high calcium loading at one site was attributed to cement plants in the sampling area. Kowalczyk (1979) reports finding a potassium to calcium ratio of 0.8 in plume aerosols collected over a cement plant near Washington, D.C. The uniqueness test for calcium shows no strong correlation between calcium and potassium, indicating the origins of the source are probably crustal. The refined source profiles that best reproduced the coarse fraction are listed in Table 5. The calculated profiles of the two crustal components follow those of Mason (1966), though the calcium concentration of 20% in the limestone factor is less than the reported value. The paint-pigment profile strongly resembles that calculated for the fine-fraction data. The only major difference is in the concentration of barium. Unlike the fine fraction, the coarse-fraction profile does not associate barium with the paint-pigment factor. The calculated sulfur concentration in the coarse-fraction sulfate factor is much less than that in the fine-fraction and there are sizable concentrations of elements such as aluminum, iron and lead not found in the fine-fraction profile. The origin of this factor is not clear. One possible explanation is that a small part of the sulfate particles in the fine fraction ended up in the coarse samples, though a correction for this possibility was made by Goulding *et al.* (1978) in the original analyses of the samples. Another possible explanation is the transformation of gaseous sulfur dioxide into sulfates on the surface of the coarse particles. Carbon soot and iron- or manganese-containing particles can catalyze the conversion of SO_2 to sulfates that would adhere to the particles in the coarse fraction.

Tables 6 and 7 summarize the average elemental concentrations along with the average observed concentrations for the fine fraction. The major elements, Al, Si, S, K, Ca, Fe and Pb, are fit very closely. The overall fit for the remaining elements is also fairly good. However, the average point-by-point errors in the reproduced data range up to a value of 350% for barium in the data without July 4th and 5th. Note that, despite the large point-by-point error, the average predicted and observed concentrations are very close, 16 and 15 ng m^{-3}, respectively. The disparity is, in part, due to the tendency of the target transformation rotation to produce profiles that represent the average composition of each source. Thus, even though the

Table 6. Summary of mass contributions (ng m^{-3}). RAPS station 112, July and August 1976. Samples from July 4th and 5th excluded. Fine fraction

Element	Motor vehicle	Sulfate	Flyash/ soil	Paint	Refuse	Total predicted	Total* observed	Average† % error
Al	23.0	21.0	170.0	0.0	0.0	230	200 ± 24	29
Si	0.0	36.0	420.0	0.0	9.0	470	450 ± 59	15
S	0.09	4570.0	62.0	2.0	0.0	4630	4360 ± 320	7
Cl	11.0	21.0	0.0	2.0	27.0	61	80 ± 9	163
K	6.0	30.0	49.0	2.4	60.0	150	150 ± 9	28
Ca	49.0	0.0	52.0	14.0	1.5	120	110 ± 10	53
Ti	0.0	12.0	8.0	46.0	0.0	66	64 ± 13	260
Mn	0.0	0.0	2.3	2.0	11.0	15	17 ± 3	122
Fe	0.0	21.0	120.0	37.0	45.0	220	220 ± 19	15
Ni	0.4	0.8	0.1	0.0	0.9	2.1	2.2 ± 0.2	106
Cu	2.6	0.2	0.0	0.0	11.0	14	15 ± 2	210
Zn	3.6	0.0	0.0	1.5	81.0	86	75 ± 8	77
Se	0.4	2.1	0.0	0.1	0.2	2.8	2.7 ± 0.2	89
Br	135.0	0.5	8.2	0.0	0.1	140	132 ± 8	27
Sr	0.4	0.2	0.5	0.1	0.0	1.2	1.1 ± 0.1	88
Ba	3.1	0.9	0.2	12.0	0.6	16	15 ± 4	353
Pb	480.0	120.0	16.0	0.0	57.0	680	720 ± 53	6

* Uncertainty is the standard deviation of the mean value.
† Average point-by-point error in predicted and measured data.

Table 7. Summary of mass contributions (ng m^{-3}). RAPS station 112, July and August 1976, including samples from July 4th and 5th. Fine fraction

Element	Motor vehicle	Sulfate	Flyash/ soil	Fireworks	Total predicted	Total* observed	Average† % error
Al	16.0	17.0	176.0	20.0	230	220 ± 31	29
Si	0.0	54.0	398.0	0.0	450	440 ± 57	12
S	0.0	4495.0	40.0	8.5	4540	4370 ± 310	5
Cl	28.0	31.0	0.9	6.2	66	85 ± 9	194
K	0.0	1.2	122.0	190.0	310	320 ± 130	11
Ca	64.0	0.1	48.0	0.1	112	112 ± 10	72
Ti	15.0	35.0	6.5	0.0	56	63 ± 13	655
Mn	8.0	2.7	2.2	1.2	14	17 ± 3	175
Fe	31.0	74.0	108.0	3.0	215	217 ± 19	49
Ni	1.0	1.1	0.1	0.1	2.3	2.3 ± 0.2	128
Cu	10.0	3.5	0.1	1.5	15	16 ± 2	261
Zn	52.0	27.0	0.0	7.9	87	78 ± 8	131
Se	0.6	2.1	0.0	0.0	2.8	2.7 ± 0.2	89
Br	138.0	0.0	7.7	0.7	146	137 ± 9	31
Sr	0.0	0.0	2.5	3.8	6.3	5.3 ± 4.0	294
Ba	7.7	5.8	2.3	4.9	21	19 ± 5	303
Pb	557.0	155.0	11.0	0.0	723	725 ± 51	2

* Uncertainty is the standard deviation of the mean value.
† Average point-by-point error in predicted and measured data.

average fit for barium is very good, the point-by-point error indicates that this element has not been well reproduced. The problem is compounded by the large number of values below the detection limits for many of the less abundant elements. The presence of source components that are not resolved can lead to the underprediction of elements. For example, a paint-pigment factor was resolved in the data without July 4th and 5th but is concealed when the data for those two days are included. The unresolved paint-pigment factor in the data with July 4th and 5th would account for the underprediction of titanium in the data that included the two days. With the addition of the two extra factors to the data without July 4th and 5th, a much better fit for the concentrations of zinc and titanium was obtained. Table 8 summarizes the mass contributions for the coarse fraction. Here, all the elements are fit well.

A comparison of the predicted and measured masses for each sample is another indicator of the quality of fit produced by the target transformation. The average deviations in the mass predictions were 16% for the fine-fraction data and 12% for the coarse. The very good fit to the mass predictions indicates that most of the undetermined elements correlate fairly closely with the measured elements. Figure 2 summarizes the

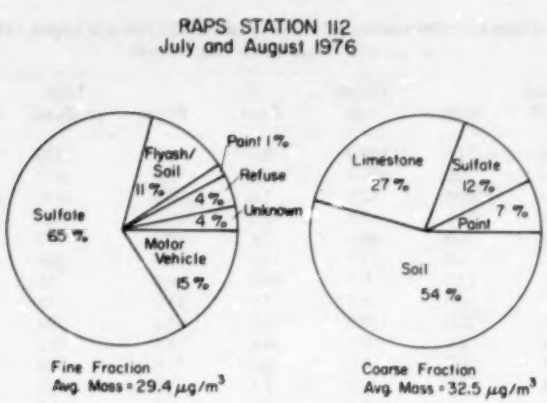


Fig. 2. Average percent contribution of each source to the total average mass. The data are the fine- and coarse-fraction samples from RAPS station 112 for the months of July and August 1976. The fine-fraction data do not include the samples from July 4th and 5th.

Table 8. Summary of mass contributions (ng m^{-3}). RAPS station 112, July and August 1976. Coarse fraction

Element	Soil	Limestone	Sulfate	Paint	Total predicted	Total* observed	Average† % error
Al	1365.0	272.0	111.0	125.0	1760	1840 ± 180	12
Si	5266.0	1359.0	0.0	0.0	6630	6400 ± 580	4
S	94.0	0.0	356.0	92.0	543	490 ± 32	35
Cl	27.0	145.0	14.0	17.0	200	210 ± 213	72
K	365.0	136.0	37.0	0.0	540	540 ± 50	9
Ca	769.0	1703.0	0.3	62.0	2540	2380 ± 141	8
Ti	0.0	14.0	0.0	320.0	330	300 ± 43	107
Mn	15.0	15.0	4.0	3.0	37	39 ± 4	37
Fe	769.0	308.0	168.0	162.0	1410	1470 ± 158	11
Ni	0.2	2.0	0.8	0.1	3.2	3.6 ± 0.4	155
Cu	0.0	5.8	3.6	0.0	9.4	8.9 ± 0.8	149
Zn	0.0	38.0	17.0	0.7	56	56 ± 6	126
Se	0.2	0.2	0.5	0.0	0.9	0.7 ± 0.04	154
Br	8.8	28.0	15.0	3.2	55	51 ± 3	51
Sr	3.8	2.4	0.7	0.3	7.2	7.9 ± 0.8	21
Ba	11.0	6.3	1.4	8.0	27	27 ± 3	53
Pb	29.0	100.0	52.0	17.0	197	193 ± 11	47

* Uncertainty is the standard deviation of the mean value.
† Average point-by-point error in predicted and measured data.

average mass contribution for each source to the total measured sample mass. The results for the fine fraction in Fig. 2 do not include the samples from July 4th and 5th. The low percentage of unaccounted sample mass is expected in this type of analysis since the regression fit calculates scaling factors so as to minimize the overall difference between the measured and predicted sample mass. However, possible uncertainties in the scaling factors of the less important sources, i.e., refuse, paint-pigment and flyash, could result in large uncertainties in the calculated concentrations of these sources. Secondary sulfate aerosol particles account for 65% of the mass of the fine-fraction data, an average concentration of about 19 μg m^{-3}. Motor vehicle emissions account for another 15%. The measured lead concentration is divided among the refuse and motor vehicle factors. Here the lead contribution is 70% from motor vehicle emissions and 10% from refuse incinerators. In the coarse fraction, the two crustal components account for 80% of the total mass.

Though carbon was not among the elements analyzed by Goulding *et al.* (1978), it is possible to estimate the carbon concentration resulting from each of the resolved sources. Watson (1979) and Pierson (1978) measured carbon concentrations in particles from motor vehicle emissions of around 500 mg g^{-1}. If this concentration were used in the present study it would result in an average carbon contribution from motor vehicles of around 2 500 ng m^{-3}, or about 9% of the average fine-fraction mass. Additional amounts of carbon may be associated with the sulfate and refuse factors. Flyash would contribute a negligible amount

of carbon (Fisher *et al.*, 1978). For the coarse fraction, Mason (1966) reports carbon concentrations in shale and limestone of 2% and 11%. Thus, the two crustal factors would represent a carbon concentration of about 2000 ng m^{-3}. If the sulfate factor in the coarse fraction indeed represents the catalyzation of SO_2 to SO_4 by carbon soot, then it would provide an additional source of carbonaceous particles.

RESULTS OF THE ANALYSIS OF SAMPLES FROM ONE WEEK AT ALL STATIONS

The data from all RAPS stations for one week were analyzed with the samples separated into fine- and coarse-fraction subsets. The results of the dimensionality tests for both fractions are given in Table 9. In both cases there are no clear indicators of the correct number of factors to be retained. The tests for the fine fraction seem to indicate either 4 or 5 factors and for the coarse fraction, 5 or 6.

Table 9. Results of dimensionality tests, 1 week, all RAPS stations

Factor	Eigenvalue	Chi square	Exner	Average % error
		Fine fraction		
1	136.00	290	0.1620	142
2	2.04	78	0.0961	94
3	0.43	47	0.0751	87
4	0.32	28	0.0547	75
5	0.10	30	0.0460	67
6	0.08	35	0.0381	64
7	0.06	35	0.0312	63
8	0.04	36	0.0250	55
9	0.03	40	0.0193	40
		Coarse fraction		
1	123.00	90	0.3118	80
2	3.46	96	0.2454	75
3	2.57	66	0.1810	66
4	1.41	53	0.1331	58
5	0.87	46	0.0917	53
6	0.36	44	0.0677	43
7	0.29	49	0.0385	38
8	0.06	50	0.0299	38
9	0.04	57	0.0211	33

The target transformation analysis of the fine fraction produced identical results for both 4 and 5 factors, indicating that 4 should be a sufficient number of factors to represent the data. The factors determined by the target transformation analysis were motor vehicle, sulfate, smelters and steel. The smelter factor was determined by the uniqueness test for the element zinc and was found to be associated with lead. Gatz (1978) noted the presence of a zinc/lead factor and attributed it to smelter located primarily in the East St. Louis and Granite City areas. The zinc factor could also arise from refuse-incinerator emissions, though such emissions are usually associated with a high chlorine concentration (Greenberg *et al.*, 1978). The steel factor was determined from the uniqueness test for iron and found to be associated with aluminum, silicon, potassium and calcium. Watson (1979) found steel- and ferromanganese-plant emissions to be associated with these elements as part of the PACS study of the Portland, Orgon airshed. The iron factor could potentially represent iron-rich flyash emissions from coal burning power plants. Gladney (1974) found iron-rich flyash particles in the size range of 1 to 3 μm at the Chalk Point electric generating station. The iron factor could also arise from entrained crustal material, though most crustal dust would be expected to be found in the coarse fraction.

The target transformation analysis of the coarse fraction again indicated the presence of two crustal sources: the first an alumino-silicate type and the second of limestone or cement origin. The uniqueness test indicated the presence of steel and sulfur sources with 5 retained factors, and a titanium source with 6 factors. Of the 17 input source profiles, there was good alignment for 3 different crustal-like sources: Soil, limestone, and coal flyash. Since there was no clear indication of the number of factors in the dimensionality tests, two separate sets of test profiles were analyzed to determine the set that best represented the data. These were: Soil, steel, limestone, sulfate and flyash, with 5 factors; and soil, steel, limestone, sulfate, flyash and paint pigment, with 6 factors. It is somewhat surprising for a flyash factor to align so well with the data in the coarse fraction since flyash particles would be expected to appear primarily in the fine fraction. It is possible that this factor represents a third crustal source, though more information would be needed to differentiate flyash and crustal material.

The scaled source vectors that best reproduced the fine and coarse data are listed in Table 10. In the fine fraction, the sulfur concentration of 19% in the sulfate factor is less than that expected for ammonium sulfate; indicating that other elements, especially carbon, may be included in this factor. In the motor vehicle factor, the high aluminum and silicon values may indicate that this factor has included small amounts of flyash or crustal material not accounted for by any of the other factors. The lead concentration of 23% is much higher than that calculated in the previous analysis, though not beyond the range cited by Watson (1979). The bromine to lead ratio is 0.19. The profile of the steel factor is similar to the steel and ferromanganese source reported by Watson (1979), though the scaled iron concentration is much less than expected.

For the coarse data, reasonable fits could be made with either 5 or 6 factors. However, the scaling factor for the paint-pigment source with 6 retained factors resulted in a titanium concentration of around 500%, indicating the extremely small contribution of this factor. A reasonable fit to the data was achieved when the paint-pigment factor was eliminated and only 5 factors retained. The scaled source vectors that best reproduced the coarse data are listed in Table 10. The paint-pigment source that was too weak to be resolved

has been absorbed into the steel and sulfate factors as manifested by the elevated titanium concentrations in these source profiles. The soil/flyash factor agrees fairly closely to that of measured coal flyash except for the somewhat higher than expected sulfur concentration. The lead and bromine concentrations are elevated in the soil, limestone and sulfate factors. The elevated concentrations could be a result of a small part of the fine particles being misdirected into the coarse fraction by the dichotomous samplers or by the possibility of motor vehicle and smelter emissions adhering to the surface of the larger particles and thus becoming a part of the coarse fraction. As was the case with the coarse-fraction data at station 112, the sulfate factor includes sizable concentrations of elements such as aluminum, calcium, titanium, iron and lead. Again, the presence of these elements could be the result of the transformation of gaseous sulfur dioxide into sulfate on the surface of the coarse particles. To more closely examine the paint-pigment factor, a separate analysis was performed on a subset of the data consisting of 28 samples that had a relatively high titanium concentration. The samples were from stations 103, 105, 106, 108 and 112, all located in the industrialized sections of the sampling area and to the north of the paint-pigment factory. A paint-pigment factor was again resolved but this time the scaling factors for some of the other sources produced physically impossible results. The wide variation in the nature of the sources between the stations seems to be inhibiting the resolution of sources. The analysis of samples from only one station, where the number of distinct sources is limited, seems to produce a superior source determination.

Tables 11 and 12 summarize the average predicted elemental concentrations for both fractions. In the fine fraction, the average percentage errors for the more abundant elements, especially aluminum and silicon, are significantly larger than those calculated in the analysis of samples from station 112. This result is probably due to the presence of an unresolved flyash or soil component that is too weak to be accounted for by this analysis. With the exception of titanium, all the major elements in the coarse fraction are fit reasonably

Table 10. Refined Source Profiles (mg g^{-1}). One week, all RAPS stations

	Fine fraction			
Element	Sulfate	Steel	Motor vehicle	Smelter
Al	2.1	28.0	22.0	30.8
Si	3.1	33.6	19.0	15.4
S	189.0	0.0	0.6	0.0
Cl	0.1	9.9	18.8	14.0
K	2.0	43.0	10.6	29.7
Ca	1.5	18.0	16.0	8.8
Ti	0.2	8.5	11.4	0.6
Mn	0.0	7.1	0.7	0.8
Fe	0.2	101.0	0.0	36.0
Cu	0.3	1.3	3.0	19.0
Zn	0.0	2.7	6.4	202.0
Se	0.1	0.1	0.6	0.9
Br	0.0	6.9	43.0	14.0
Ba	0.0	2.9	4.7	0.8
Pb	1.1	0.0	227.0	156.0

	Coarse fraction				
Element	Soil	Steel	Limestone	Sulfate	Soil/ flyash
Al	70.0	40.3	24.4	12.4	140.0
Si	294.0	0.0	131.0	0.0	150.0
S	0.3	13.3	6.2	131.0	0.2
Cl	0.0	99.0	13.0	0.0	18.0
K	23.0	1.0	13.3	12.1	21.7
Ca	54.0	62.0	240.0	40.0	19.0
Ti	0.0	10.0	13.0	22.6	1.4
Mn	1.1	23.0	0.0	1.7	0.0
Fe	55.0	670.0	1.5	25.5	33.0
Cu	0.2	0.2	0.0	3.2	1.1
Zn	1.7	35.0	2.1	4.8	2.5
Se	0.1	0.1	0.0	0.0	0.1
Br	0.9	0.6	1.8	3.4	2.0
Ba	0.3	0.0	1.2	1.8	3.7
Pb	2.8	13.0	9.0	15.0	2.7

Table 11. Summary of mass contributions (ng m^{-3}). One week, all RAPS stations

	Fine fraction						
Element	Sulfate	Steel	Motor vehicle	Smelter	Total predicted	Total* observed	Average† % error
Al	40.0	41.0	28.0	18.0	127	117 ± 5	62
Si	59.0	49.0	24.0	9.0	142	151 ± 9	64
S	3600.0	0.0	0.8	0.0	3600	3560 ± 150	1
Cl	1.9	14.0	24.0	8.3	49	52 ± 6	169
K	38.0	63.0	14.0	18.0	132	137 ± 10	32
Ca	29.0	26.0	20.0	5.2	81	82 ± 5	61
Ti	4.0	12.0	15.0	0.3	31	39 ± 7	195
Mn	0.0	10.0	0.8	0.5	12	12 ± 1	119
Fe	3.8	149.0	0.0	21.0	173	169 ± 13	35
Cu	5.5	1.9	3.8	11.0	23	24 ± 4	208
Zn	0.0	4.0	8.2	120.0	131	129 ± 16	23
Se	1.9	0.1	0.8	0.5	3.3	3.5 ± 0.3	88
Br	0.0	10.0	55.0	8.3	73	68 ± 5	45
Ba	0.0	4.2	6.0	0.5	11	11 ± 3	224
Pb	21.0	0.0	290.0	92.0	403	407 ± 25	3

* Uncertainty is the standard deviation of the mean value.
† Average point-by-point error in predicted and measured data.

Table 12. Summary of mass contributions (ng m^{-3}). One week, all RAPS stations

	Coarse fraction							
Element	Soil	Steel	Limestone	Sulfate	Soil/ flyash	Total predicted	Total* observed	Average† % error
Al	330.0	31.0	140.0	20.0	230.0	747	753 ± 42	2
Si	1390.0	0.0	740.0	0.0	240.0	2380	2390 ± 160	2
S	1.2	10.0	35.0	210.0	0.3	256	259 ± 15	8
Cl	0.0	76.0	74.0	0.0	29.0	179	193 ± 17	83
K	110.0	0.7	75.0	19.0	35.0	239	237 ± 15	19
Ca	260.0	48.0	1360.0	64.0	31.0	1750	1650 ± 120	6
Ti	0.0	7.7	74.0	36.0	2.3	120	130 ± 21	170
Mn	5.2	18.0	0.0	2.7	0.0	26	24 ± 3	53
Fe	260.0	520.0	8.5	41.0	53.0	880	880 ± 80	2
Cu	0.7	0.1	0.0	5.1	1.8	7.7	7.8 ± 0.7	135
Zn	8.1	27.0	12.0	7.7	4.0	59	52 ± 5	110
Se	0.2	0.1	0.0	0.0	0.2	0.5	0.4 ± 0.03	90
Br	4.1	0.4	10.0	5.4	3.2	23	21 ± 2	76
Ba	1.3	0.0	6.8	2.9	6.0	17	17 ± 2	166
Pb	13.0	10.0	51.0	24.0	4.4	103	100 ± 9	71

* Uncertainty is the standard deviation of the mean value.
† Average point-by-point error in predicted and measured data.

well. The presence of the unresolved paint-pigment factor is the probable cause for the underprediction of the titanium concentration.

Figure 3 and 4 plot the mass concentration for each source at each station. As in the previous analysis, the main contributor to the fine fraction is the sulfate aerosol. The smelter component is most important at stations 103 and 108; these stations are located to the north of the lead and zinc smelters in East St. Louis and Granite City. The contribution of the steel factor in the fine and coarse fractions is significant at only four stations: 103, 105, 106, and 108. These stations are located in the heavily industrialized sections of the sampling area and in close proximity to the iron works and foundries. The average deviations in the mass predictions are 32 % in the fine fraction and 34 % in the coarse; significantly poorer than those obtained for the

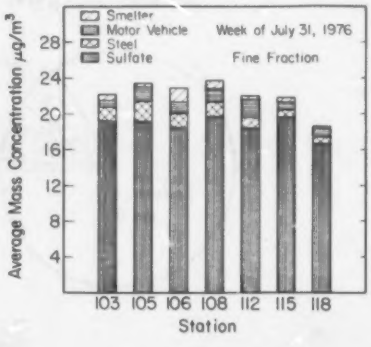


Fig. 3. A comparison of the calculated mass concentrations of the 4 source components in the fine-fraction data from all RAPS stations. The samples are from the week beginning July 31, 1976.

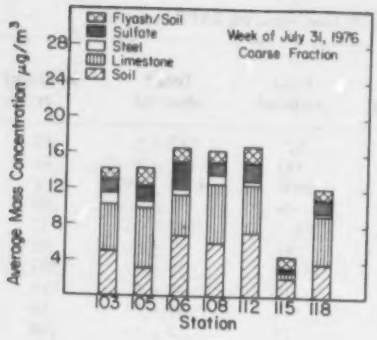


Fig. 4. A comparison of the calculated mass concentrations of the 5 source components in the coarse-fraction data from all RAPS stations. The samples are from the week beginning July 31, 1976.

data from station 112. The poorer fit probably results from the presence of unresolved sources.

Figure 5 summarizes the average mass contribution of each source for the fine and coarse fractions. The predicted motor vehicle concentration of 6% is lower than in the analysis of the station 112 data. The measured lead concentration is apportioned among two factors: motor vehicle emissions account for about 72% of the lead concentration while smelters contribute about 23%. In the coarse fraction, the two crustal components account for 70% of the total mass. The soil/flyash component contributes an additional 10%.

If the concentration of carbon in motor vehicle emissions is again assumed to be 500 mg g^{-1} then the ambient carbon concentration from this source would be about 700 ng m^{-3}. In the coarse fraction, the estimated carbon contribution from the crustal sources would be about 1000 ng m^{-3}. However, the large average percentage error in the measured versus predicted sample masses may indicate that the undetermined elements do not correlate very well with the analyzed elements and attempts to estimate the contribution of the unmeasured elements would be difficult and the reliability of results highly uncertain.

CONCLUSIONS

Target transformation factor analysis has been used to resolve sources of airborne particulate matter in samples collected as part of the RAPS study of St. Louis, MO. For the samples from station 112, a source profile for fireworks was resolved in the fine-fraction data, even though this factor was present in only 3 of the 100 samples. The presence of the fireworks factor, however, was found to conceal the presence of other, weaker, sources. Additional factors were resolved when the samples from July 4th and 5th were excluded. Five factors were needed to represent the fine-fraction data without the samples from July 4th and 5th: sulfate, motor vehicles, flyash/soil, paint-pigment, and refuse incinerators. A source profile for coal-burning power plant emissions was tentatively identified, though distinguishing crustal material from coal flyash would be more certain if data for additional elements were available. Four factors were needed to represent the coarse-fraction data: limestone, sulfate, and paint-pigment.

Overall, the analysis of the samples from all stations for one week resulted in a poorer resolution of sources. The nature of the sources contributing to each station are sufficiently different so as to preclude a resolution of all but the most important sources. Only 4 factors were resolved from the fine-fraction data: sulfate, steel, motor vehicles, and smelters. The higher than expected aluminum and silicon concentrations in the motor vehicle factor indicate that this factor represents an amalgamation of different sources that could not be re-

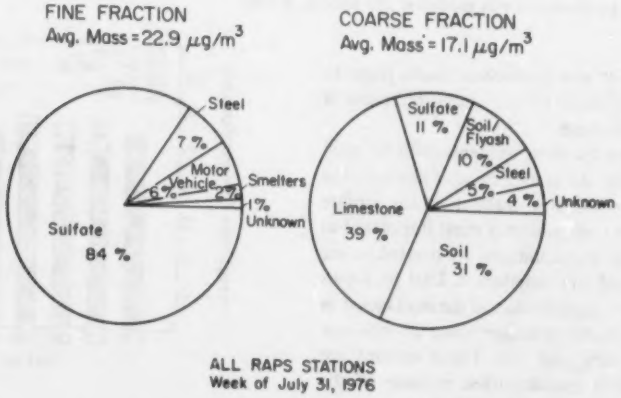


Fig. 5. Average percent contribution of each source to the total average mass. The data are the fine- and coarse-fraction samples from all RAPS stations for the 7 days beginning July 31, 1976.

solved. Five factors were resolved for the coarse fraction: soil, steel, limestone, sulfate, and soil/flyash. The presence of a paint-pigment factor was indicated by the uniqueness test but was too weak to be clearly resolved. The presence of 3 crustal sources could not be explained. The average percentage error in the calculated and measured mass of each sample was significantly poorer for the data from all RAPS stations for one week.

Acknowledgements—This work was supported in part by the Environmental Protection Agency (USEPA PO D6004NAEX). One of us (D.J.A.) wishes to express his appreciation to the Public Health Service, Department of Health, Education, and Welfare, National Research Award Number 5 T32 ES07001-04, for their support.

REFERENCES

Alpert D. J. and Hopke P. K. (1980a) Modified factor analysis of selected RAPS aerosol data. Final report to the Environmental Protection Agency, Order Number D6004NAEX.

Alpert D. J. and Hopke P. K. (1980b) A quantitative determination of sources in the Boston urban aerosol. *Atmospheric Environment* (in press).

Clifford A. A. (1973) *Multivariate Error Analysis*, pp. 71–75. Applied Science, Ltd., London.

Commission on Natural Resources, National Research Council (1979) *Controlling Airborne Particles*. National Academy of Sciences, Washington, D.C.

Currie L. A., Kunen S. M., Voorhees K. J., Murphy R. B. and Koch W. F. (1978) Analysis of carbonaceous particulates and characterization of their sources by low-level radiocarbon counting and pyrolysis/gas chromatography/mass spectrometry. In *Proc. Conf. Carbonaceous Particles in the Atmosphere*, Lawrence Berkeley Laboratory Report LBL-9037, CONF-7803101, UC-11.

Daisey J. M., Hawryluk I., Kneip T. J. and Mukal F. (1978) Mutagenic activity in organic fractions of airborne particulate matter. In *Proc. Conf. Carbonaceous Particles in the Atmosphere*, Lawrence Berkeley Laboratory Report LBL-9037, CONF-7803101, UC-11.

Dzubay T. G., Stevens R. K. and Richards L. W. (1979) Composition of aerosols over Los Angeles freeways. *Atmospheric Environment* **13**, 653–659.

Dzubay T. G. (1980) Chemical element balance method applied to dichotomous sampler data. *Ann. N. Y. Acad. Sci.* **338**, 126–146.

Exner O. (1966) Additive physical properties, *Collect. Czech. Chem. Commun.* **31**, 3222–3253.

Fisher G. L., Chrisp C. E. and Hayes T. L. (1978) Carbonaceous particles in coal fly ash. In *Proc. Conf. Carbonaceous Particles in the Atmosphere*, Lawrence Berkeley Laboratory Report LBL-9037, CONF-7803101, UC-11.

Gaarenstroom P. D., Perone S. P. and Moyers J. L. (1977) Application of pattern recognition and factor analysis for characterization of atmospheric particulate composition in southwest desert atmosphere. *Envir. Sci. Technol.* **11**, 795–800.

Gatz D. F. (1978) Identification of aerosol sources in the St. Louis area using factor analysis. *J. appl. Met.* **17**, 600–608.

Gladney E. S. (1974) Trace elemental emissions from coal-fired power plants: A study of the Chalk Point Electric Generating Station. PhD Thesis, University of Maryland.

Goulding F. S., Jaklevic J. M. and Loo B. W. (1978) Aerosol analysis for the Regional Air Pollution Study—Interim report. EAP-600/4-78-034, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Greenberg R. R., Gordon G. E. and Zoller W. H. (1978) Composition of particles from the Nicosia Municipal Incinerator. *Envir. Sci. Technol.* **12**, 1329–1332.

Hopke P. K., Gladney E. S., Gordon G. E., Zoller W. H. and Jones A. G. (1976) The use of multivariate analysis to identify sources of selected elements in the Boston urban aerosol. *Atmospheric Environment* **10**, 1015–1025.

Hopke P. K., Lamb R. E. and Natusch D. F. S. (1980) Multielemental characterisation of urban roadway dust. *Envir. Sci. Technol.* **14**, 164–172.

Kowalczyk J. F., Core J. E. and Cooper J. A. (1979) The true sources of Portland's suspended particulate. Presented to the Pacific Northwest International Section, Air Pollution Control Association, November 1979, Paper Number 79-10.5.

Kowalczyk G. S. (1979) Concentration and sources of elements on Washington, D.C. area atmospheric particles. PhD Thesis, University of Maryland.

Lewis C. W. and Macias E. S. Composition of size-fractionated aerosol in Charleston, West Virginia. *Atmospheric Environment* **14**, 185–194.

Lodge J. P., Jr., Bien, G. S. and Suess H. E. (1960) The carbon-14 contents of urban airborne particulate matter. *Int. J. Air Pollut.* **2**, 309–312.

Loo B. W., Jaklevic, J. M. and Goulding, F. S. (1976) Dichotomous virtual impactors for large scale monitoring of airborne particulate matter. In *Fine Particles*, (Edited by B. Y. H. Liu), pp. 311–350. Academic Press, New York.

Malinowski E. R. and Howrey D. G. (1980) *Factor Analysis in Chemistry*. John Wiley and Sons, Inc., New York

Mason B. (1966) *Principles of Geochemistry*, pp. 153–181. John Wiley and Sons, Inc., New York.

Miller M. S., Friedlander S. K. and Hidy G. M. (1972) A chemical element balance for the Pasadena aerosol. *J. Colloid Interface Sci.* **39**, 165–176.

Nelson E. (1979) Regional air pollution study: Dichotomous aerosol sampling system, EPA-297 310, U.S. Environmental Protection Agency, Research Triangle Park, NC.

Pierson W. R. (1978) Particulate organic matter and total carbon from vehicles on the road. In *Proc. Conf. Carbonaceous Particles in the Atmosphere*, Lawrence Berkeley Laboratory Report LBL-9037, CONF-7803101, UC-11.

Rheingrover S. W. (1977) A statistical model for titanium pollution transport and dispersion in the atmosphere of St. Louis. M. S. Thesis, Florida State University.

Watson J. G., Jr. (1979) Chemical element balance receptor methodology for the sources of fine and total suspended particulate matter in Portland, Oregon. PhD Thesis, Oregon Graduate Center.

Weiner P. H., Malinowski E. R., and Levinstone A. R. (1970) Factor analysis of solvent shifts in proton magnetic resonance. *J. Phys. Chem.* **74**, 4537–4542.